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Manufacturing of Annular Targets Made of LEU Foil Coated with Electrodeposited Nickel

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Abstract

The results achieved in the development of an annular target manufacturing technology for Mo-99 radioisotope production, with targets containing a low enriched uranium LEU foil, are presented in this paper. The paper describes the annular targets manufacture comprising the uranium foil and a thin layer of nickel obtained by electroplating on the surface of the uranium, resulting in Ni deposits with thickness of 15 to 25 microns. It was necessary to determine the potential to avoid the presence of uranium oxide and also to determine the intensity (amperage) at which its formation begins. The results obtained, showed that the mass of Ni deposited on the uranium foil is proportional to the electroplating time and the current intensity. Ultrasonic Tests and Screen Electronic Microscopy revealed good bonding and homogeneity of the nickel deposit. The Ni layer had a high hardness and a relatively small grain size with an orientation in the direction of the electrical current flow.

Keywords: Electrodeposition, Uranium, Nickel, Annular Target, Fission Mo

1. Introduction

The Chilean Nuclear Energy Commission, CCHEN, produces amongst other radio isotopes for nuclear medicine ^{99m}Tc, ^{131}I, ^{192}Ir, ^{153}Sm in its research reactor RECH-1. The most used radioisotope worldwide for medical procedures is metastable Technetium–99 (^{99m}Tc), a product of the radioactive disintegration chain of ^{99}Mo. This radioisotope is applied in medical diagnosis procedures for diverse pathologies, concentrating around 80% of all gamma cameras images obtained in nuclear medicine procedures. This 80% correspond to more than 25 millions of studies per year, Goldman et al. (2008). In Chile is applied to more than 90% of the nuclear medicine studies (about 200.000 patients). At the RECH-1in Santiago, the ^{99m}Tc is produced by the neutronic activation of molybdenum,

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while in the rest of the country; the ^{99m}Tc is produced by imported generators, Schrader et al (2007). A Technological Development Program in progress at CCHEN considers the production of ^{99}Mo based on the irradiation of annular targets containing a LEU uranium foil inside.

CCHEN's Nuclear Materials Department have joined efforts to overcome the development challenge for the annular targets manufacturing technology, including the design and manufacture of equipment, tooling and materials for the assembly and disassembly of the annular target after its irradiation in the reactor. The annular target comprises two concentric tubes made of Al-3003 AISI-SAE alloy, and between these tubes, into a properly machined annular region, a thin foil of metallic LEU is placed and encapsulated, on all its sides, by a nickel deposit which acts as containment barrier for the fission products produced during the irradiation, Schrader et al (2008), Lisboa (2009), ANL. The assembly is showed in Fig. 1.

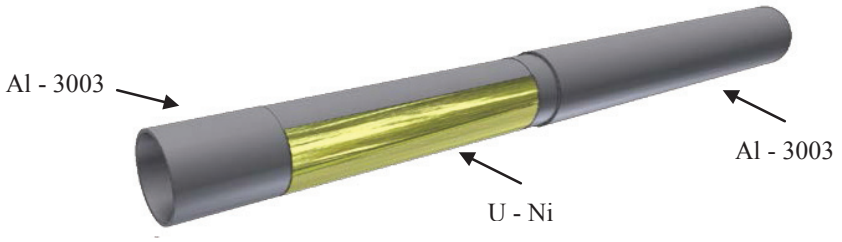

Fig.1. Cut view of the assembly of U-Ni and Al-3003.

The main difficulty after irradiation is the extraction of the uranium foil from the target. This is attributed to the bonding between the uranium foil and the structural aluminum tubes that is originated by the ions mixture caused by the fission products from the uranium foil. Therefore nickel is placed between the Al tubes and the uranium foil and thus used as a barrier to avoid the escape of fission products. The thickness of the barrier is determined by the desired range of recovery of fission products. Maximum recovery distance is about 7 microns; however, to provide a safety range, a layer with 15 microns thickness is selected, which corresponds to twice the maximum distance. This layer should cover completely the uranium foil to avoid the possibility of localized bonding with the Al tube wall. This material should have a uniform thickness to ensure a proper fit during the assembly and has shown proper heat dissipation during irradiation, Smaga et al. (1997). Targets that use the nickel barrier have shown good behavior under irradiation, from the point of view of heat transfer and also to prevent bonding between the uranium foil and Al tube wall during irradiation, Conner et al. (1999), Conner et al. (2000).

The development of the manufacturing technologies for annular target at CCHEN is focused on the production of uranium foil with thickness about 120 microns with a nickel electroplating on this foil. This nickel coating should be characterized to comply with the specifications for an effective barrier to fission products. Accordingly, the Ni electrodeposition process for the uranium foil is intended to solve certain technical details that happen when thin nickel films are used, and therefore to improve the production and the quality of the annular targets.

2. Experimental set-up/ methodology

The development of this work started with the casting of a uranium metal ingot obtained by induction melting in a multipurpose chamber with controlled atmosphere. In the melting process a high alumina crucible was used and the liquid metal was poured into a closed graphite mold, integrally mounted with the crucible in a semi – Durville assembly.

The uranium ingot must be protected from oxidation during the hot rolling process. For this purpose, cover and frame of low carbon steel was used as a sealing cladding. Considering that the aim of this stage is to obtain a uranium foil, the steel surfaces in contact with the uranium ingot were coated with an emulsion of yttrium oxide (Yttria) and ethanol, and thus preventing bonding and interaction between the uranium ingot and steel. Once applied this protective coating to the surfaces, the set was assembled and fixed by TIG welding.

Before the hot rolling process, the assembly was annealed 1 hour at 630 °C in air. Thickness reductions of 5 % were applied in the first four rolling steps and the following reductions steps were done applying 10% of reduction in each step, with intermediate annealing of 10 minutes between each reduction step.

During the hot rolling process and every 5 reduction steps, the assemblies were removed from the furnace and radiographed for metrological control. The rolling deformation achieved values between 94 % and 96% of the total reduction in the thickness of steel - uranium - steel assemblies. For cold rolling it was necessary to perform a surface cleaning to the uranium foils in order to remove the coating of yttrium oxide, and then the uranium foils were encapsulated into envelopes of 304 stainless steel type, primarily for protection of the work rolls and to avoid its surface contamination with uranium. The reductions applied at this stage were under 3% in each rolling step, reaching values between 7% and 14% of total reduction in thickness. Uranium foils were characterized mechanically by tensile tests according to ASTM E -345standard for metal foil.

An electrodeposition system was built comprising two cells containing the electrolyte; a first cell to heat the electrolyte and pH control of the solution, and the second cell for electrodeposition, as shown in Fig. 2. The heating of the solution was performed by recirculating warm water in a coil placed in the first cell. The stirring and homogeneity of the solution was accomplished by recirculating the solution between the two stainless steel cells.

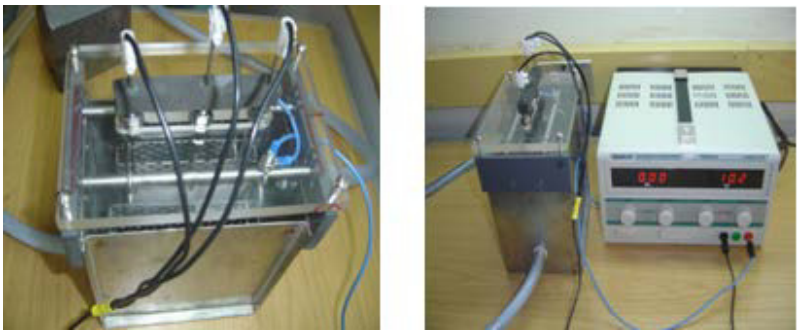

Fig.2. Electrodeposition system

The electrolyte solution was prepared with 250 g/l nickel sulphate; 60 g/l nickel chloride, 40 g/l of boric acid and deionized water. Besides, 2 ml/l of Omega additive was added to the solution to improve the ductility and to give a semi glossy aspect to nickel deposit, and 4 ml/l of wetting agent was added to the Ni solution to reduce its surface tension to avoid the occurrence of porosity in the nickel deposit. Nickel sulfate is the primary source of nickel ions for the electrolytic solution, while nickel chloride contributes to the dissolution of the activated forms of nickel, and boric acid stabilizes the pH of the solution.

Deposits characterization was performed by thickness measurement using a linear comparator and also by means of optical microscopy, scanning electron microscopy, and ultrasonic scanning.

3. Results and discussion

3.1. Uranium Foil Fabrication

The dimensions of the ingots obtained are presented in Table 1. The ingots were cut to predetermined dimensions according to the final dimensions required for the uranium foil. Furthermore, the results of hot rolling, indicating the percentage of total reduction applied in each case, are included.

Table 1. Dimensions of uranium samples As-cast and hot rolled.

	Dimensions of As-cast coupons			Dimensions of hot rolled samples				
Identification	Thickness	Wide	Length	T	Reduction	Thickness	Wide	Length
	mm	mm	mm	°C	%	mm	mm	mm
FUN01 (U)	4.94	21.56	42.03	630	95.93	0.125	46	701
FUN02 (U)	6.02	30.07	39.52	630	94.52	0.153	37	876

Fig.3. Uranium Foils.

The encapsulation technique used to obtain the uranium foil gave good results, and the yttrium oxide coating effectively inhibited bonding between the uranium and the low carbon steel cladding. Fig. 3 shows open assemblies and uranium foils sized and ready for assembling in annular targets. Before assembling, these foils must be chemically cleaned with nitric acid in order to remove both the yttrium oxide and any surface uranium oxide layer.

The behavior of the uranium metal during rolling was evaluated using X-Ray film, whose results showed a high elongation in both foils, increasing strongly for reductions higher than 95% on FUN-01 foil, rolled longitudinally, when compared with the FUN-02 which was rolled transversally. These foils were submitted to a tensile test and the tests results are shown in Fig. 4. The strength obtained in the foil FUN-01 FUN was 392 MPa compared with that obtained in the FUN-02, 479 MPa. These properties explains the difference in elongation of test samples: 2394% for FUN-01 and 1711% in the FUN-02. Considering the influence of the rolling direction in the ultimate strength of each foil, it can be inferred that for the uranium foil transversally rolled, an increase in the strength of the foil was obtained.

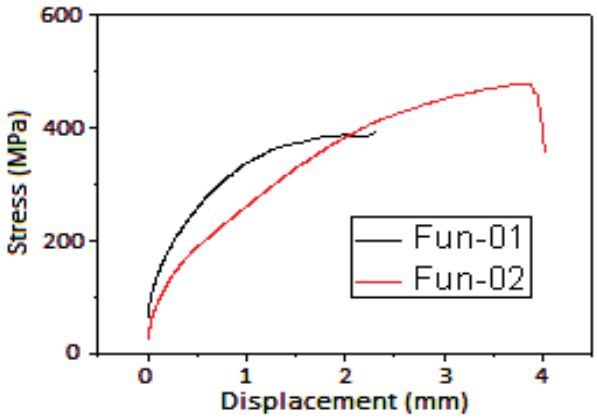

Fig.4. Tensile Test of uranium foils.

3.2. Nickel electrodeposition on the uranium foil

Uranium is very susceptible to oxidation, and this well-known fact made necessary to perform cyclic voltammetry to determine the potential in which uranium metal is obtained without the presence of an oxide layer, additionally is possible to determine the electrical current where this oxide begins to form. The starting point for the nickel deposit is (-0.660, -0.01006 V) potential and intensity of electric current, from which is possible to know that the electric current intensity required to start the deposition of Ni in the uranium surface is 0.5 A. The conditions used for the determination of the curve in Fig. 5 were pH 4.0 and a temperature of 42 °C. To activate the oxide formation a low potential was used and thus assuring a nickel deposit over a clean surface.

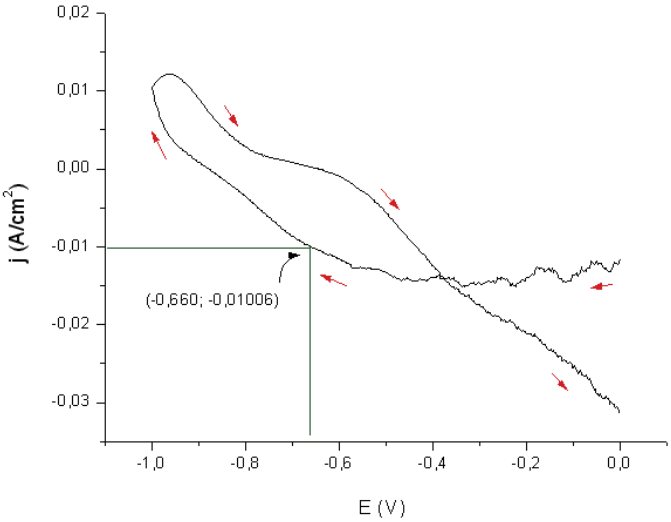

Fig.5. Cyclic voltammetry in U-Ni system

Table 2 presents the results of the experiments carried out on natural uranium foils. According to the sequence shown in Fig. 6, the deposited mass is close to the predicted value by Faraday's Law. The Omega additive effectively improved the appearance of the deposit, producing clean and shiny surfaces as shown in the last picture of Fig. 6. The surface distribution of the deposit exhibited acceptable homogeneity and the higher thickness were at the bottom of the foil, indicating that the ion flow had a trajectory according to the expected, Kanani (2005).

Table 2. Results of Ni electrodeposition over U foil.

ID	I (A)	t (min)	M (g)	M^1 (g)	e^2 (μm)
11.2	3.0	30	1.5	1.6	18.4
12.1	3.0	40	2.0	2.2	24.6
10.1	3.2	30	1.6	1.8	19.7
12.2	3.2	40	1.9	2.3	26.2
11.1	3.4	20	1.4	1.2	13.9
8	3.4	30	1.8	1.9	20.9
12.3	3.4	40	1.9	2.5	27.9

M^1= Calculated mass Faraday's Law
e^2 = Calculated thickness Faraday's Law

Fig. 6. Pictures sequence of electrodeposition of Ni over U foil.

The thickness measurement of the nickel deposit for the foil 11.2 revealed the distribution profile shown in Fig. 7. For 24 points measured over 50 cm^2 of surface, the average thickness value was 32.3 microns. The micrograph of Fig. 8 showed no surface porosity in the Ni deposit and the occurrence of defects caused by the presence of hydrogen was minimized by the addition of a wetting agent that modified the surface tension between the substrate and the electrolyte, as indicated by Kanani (2005). It also showed a thickness of 135 microns for the uranium foil with a nickel deposit thickness of 34.8 microns.

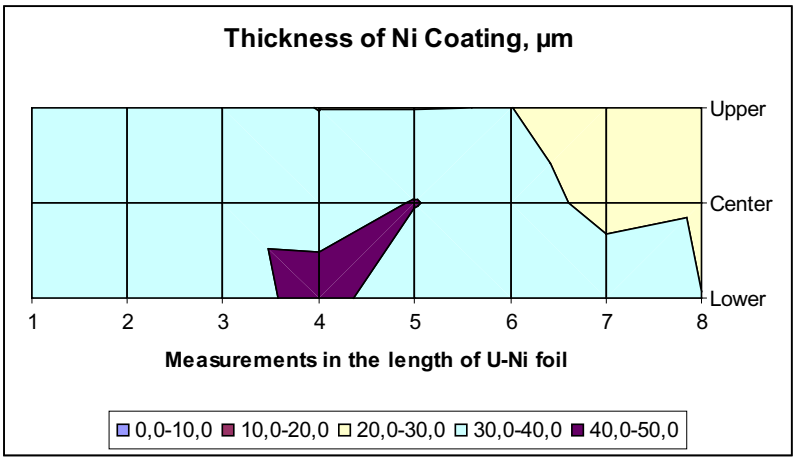

Fig. 7. Thickness Distribution for nickel deposit over uranium foils (foil 11.2)

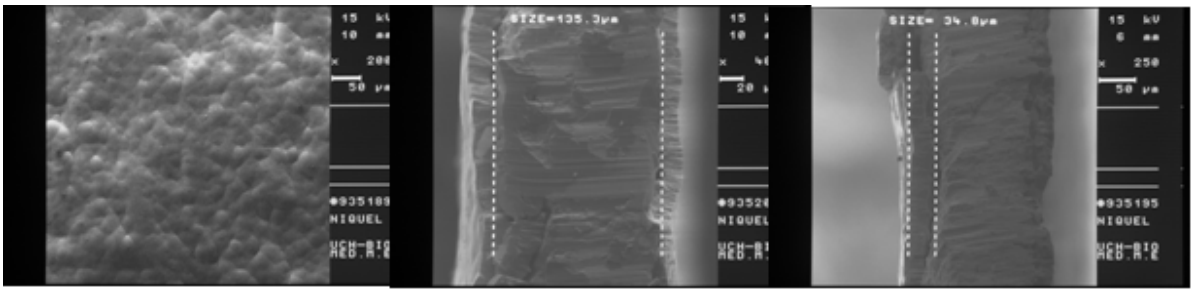

Fig. 8. Micrographs of surface and cross section of uranium foil.

Ultrasonic testing scanning inspection was performed to know the bonding integrity of the nickel deposit. Fig. 9 corresponds to a C-scan image of a linear scan for the uranium foil 12.2, where the sample exhibited a continuous signal indicating that a qualitatively acceptable bonding of the deposit over the uranium foil.

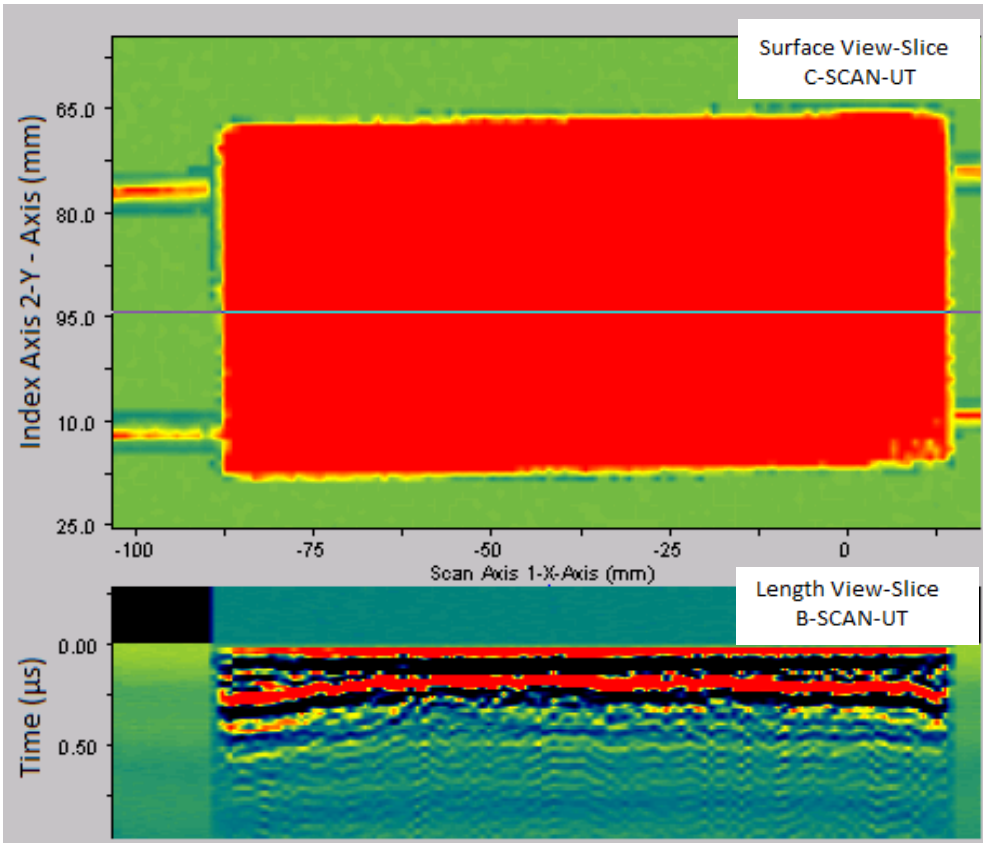

Fig. 9. C-Scan image of UT applied to 12.2 uranium foil.

Finally, Fig. 10 shows an annular target assembly with a LEU foil inside, including engraved identification and surface finishing.

Fig. 10. Annular Target finished.

4. Conclusions

The closed semi-dourville mold design, integrally mounted with high alumina crucible allows the production of ingots with a good surface quality.

The encapsulation technique with low carbon steel cladding is a very effective way to produce uranium foil

The electrolytic process demonstrates to be a suitable process for the U-Ni system, obtaining a homogenous nickel deposit with good adhesion and free of porosity.

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